Magnetic Properties of Ultra Thin Epitaxial Fe Films on GaAs(001)

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Abstract

The magnetic properties of epitaxial Fe films on GaAs in the range of the first few monolayers have been the subject of a considerable number of investigations in recent years. The absence of magnetic signatures at room temperature has been

attributed to the existence of a magnetic 'dead' layer as well as superparamagnetism. By examining the temperature dependence of the magnetic linear dichroism of the Fe core level photoelectrons, we found that these 'non-ferromagnetic' layers had in fact a Curie temperature, T_c, substantially lower than room temperature, e.g., a T_c of about 240K for thin films of a nominal thickness of 0.9 nm. The values of Curie temperature were sensitive to the initial GaAs substrate conditions and the thickness of the Fe over-layer with a layer of thickness of 1.25 nm showing a T_c above room temperature. The data suggest that the ultrathin Fe films on GaAs(001) are ferromagnetic, although a weaker exchange interaction in the films lead to a substantial reduction in Curie temperature.

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Current studies of ultra thin magnetic films on semiconductor substrates are largely motivated by possible applications in spin injection devices [1,2,3,4]. Recent demonstrations of spin-coherent transport over large distances in n-type semiconductors [5] and the persistence of spin coherence over a sizeable time scale [6], albeit at a low temperature, are clearly very encouraging. Since the early work of Waldrop and co-workers [7] there have been intensive investigations on the characteristics of Fe thin films on GaAs substrates, as a typical epitaxial magnetic metal- semiconductor system. A variety of different features has been revealed: the interfacial magnetism and its dependence on the intermixing of Fe,Ga and As on the one hand and on the morphology of the deposited Fe on the other hand. Of relevance to the present work, we note that it was proposed that As diffusion into the Fe layer and the formation of non-ferromagnetic $FeAs_2$ like compounds resulted in a magnetically dead interfacial region; above a critical thickness the magnetisation of the Fe layer would depend exponentially on the distance to the interface, correlated with the diffusion of As [8]. Similarly, interdiffusion and compound formation have been highlighted in recent studies by Filipe *et al* [9]. The exponential increase in the magnetisation observed was interpreted as an intermediate phase of ternary pseudo-cubic hexagonal FeGaAs, sandwiched between magnetically dead Fe at the interface and pure bulk-like Fe at a thickness of about 20 nm. More recently, in a study using the technique of magneto optical Kerr effect (MOKE), the delayed onset of ferromagnetic behaviour in ultra thin Fe was attributed to Fe clusters forming a superparamagnetic phase which became ferromagnetic as the clusters coalesce at higher Fe coverage [10]. Furthermore, a temperature-dependent measurement of magneto resistance of the ultrathin Fe films was attributed to the existence of such a superparamagnetic phase [11].

Whilst recognising that the magnetic behaviour of ultrathin Fe layers is also sensitive to both GaAs substrate surface morphology [12] and surface stoichiometry [13], this Letter reports on the temperature dependent magnetic state of Fe on GaAs(001) through a study of the magnetic dichroism of the Fe 2p and 3p core level photoelectrons with linearly polarised radiation.

In recent years X-ray magnetic linear dichroism (XMLD) has been well established as a surface-sensitive, element specific probe of sample magnetisation [14,15,16], which is highly suitable for the study of magnetic properties of ultrathin films of a few atomic layers. Although experimentally a relatively straight-forward measurement, the theory of XMLD is complex and for quantitative analysis a simple and effective model has been developed [16], in the case of 3p states, which has established a proportional relationship between linear dichroism asymmetry and the core-level exchange splitting, which in turn is assumed to be directly dependent on the valence state exchange splitting and the magnetic moment in the plane of the surface[17]. Therefore it is sufficient in the present study to consider the dichroism asymmetry as a direct measure of the magnetisation of the specimens.

Experiments were carried out with base pressure better than 3×10^{-10} mbar in the Spin Chamber on Beamline 7.0 at the Advanced Light Source, Berkeley [18,19]. The Fe/GaAs samples were prepared *in situ* for the dichroism study, using As capped n-type GaAs epilayers grown on n^+ singular and vicinal GaAs(001) substrates. The As capping layers were removed by thermal desorption prior to Fe deposition. For the clean substrates used in this study, a value of Ga to As ratio of about 1.06 was obtained. In a similar experimental configuration as that previously reported [16], dichroism spectra of the Fe 2p and Fe 3p states were taken using p polarised 45° incidence photons and normally emitted photoelectrons. A magnetising field was

applied perpendicular to the incidence plane of the light and the surface normal with the [110] direction of the sample orientated parallel to the field direction. The data were taken with the sample in a remnant magnetisation state.

As an example, Figure 1 shows a typical dichroism result of a 2.7 nm thick Fe film on singular GaAs (001) substrate obtained with 810 eV photons. The upper part of Figure 1 displays the raw data of the two Fe 2p core level spectra acquired with the Fe film in opposite remnant states along the easy axis in the [110] direction. Both core levels, the Fe $2p_{1/2}$ and the Fe $2p_{3/2}$, exhibit dichroic features.

The magnetic asymmetry, A_M, of the core level spectra can be calculated using:

$$A_M = \frac{I^\uparrow - I^\downarrow}{I^\uparrow + I^\downarrow} \qquad (1)$$

where $I^\uparrow$ and $I^\downarrow$ are the measured X-ray photoelectron spectroscopy (XPS) intensities, with Shirley background [20] removed, for the sample magnetized parallel, $\uparrow$, and antiparallel, $\downarrow$, to the [110] direction respectively.

The lower part of Figure 1 shows the asymmetry spectrum of the two opposite remnant states. The overall features of the spectrum agree well with those of the Fe 2p dichroism results in the literature obtained in a similar experimental geometry. For example the spectral features are similar to those observed previously by Rose *et al* [21] and Waddill *et al* [22]. Furthermore, we have found that Fe 3p dichroism results of the same sample (shown in Figure 2) are of very similar magnitude. The Fe 3p states have been used for the temperature dependent study as the proportional relationship between the XMLD asymmetry and magnetisation has been previously tested for the 3p states[16].

In Figure 3, the peak-to-peak magnetic asymmetries ('total asymmetry'), A_{TOT} as indicated in Figure 2, obtained from Fe 3p photoelectrons are displayed as a function of temperature for 0.9 nm Fe grown on a singular (unfilled circles) and a vicinal (filled circles) GaAs(001) substrate. The vicinal substrate has an off-cut of 3 degree towards the [110] direction. Both samples were grown side-by-side for better comparison and the data taken alternatively as the samples were being warmed up slowly. The results clearly show that both 0.9 nm Fe films are ferromagnetic with a similar Curie temperature of about 240K. It is also interesting to note that, the value of the magnetic asymmetry for the Fe on the vicinal substrate is about 8% higher in the temperature range studied, indicating a somewhat higher magnetisation at the same temperature. This appears to be consistent with our earlier work [12] that, in the ultrathin region, the magnetic characteristics appeared to be enhanced for Fe grown on vicinal substrates.

The value of the Curie temperature has also been found to be very sensitive to surface chemistry as well as surface morphology. For example, we have found that poorly de-capped or contaminated substrates lead to the Fe overlayers having substantially lower values of Curie temperature.

As a technique for probing magnetisation, XMLD would enable us to investigate the temperature dependent remnant states of these ultrathin films thereby gaining further understanding of their magnetic characteristics. However for the present set of results, both the statistical quality of the data and the unavailability of temperatures below 180K have limited a more quantitative analysis. Nevertheless, to illustrate the potential of temperature dependent XMLD studies, we shall compare our results with a Weiss-type theoretical model [23]. In the inset of Figure 3, we have reproduced the theoretical curves of normalised spontaneous magnetisation as a

function of reduced temperature [23]. For clarity, the experimental data of bulk Fe and Ni in the original diagram have not been included. Due to our limited temperature range, the information on the zero temperature magnetisation, M_0, is lacking. Unfortunately, fitting to the different values of total momentum quantum number, J, is sensitive to M_0. However we note that for thicker films the magnetic properties as measured by spin polarised photoelectron spectroscopy displayed bulk-like behaviour [13] and the values of Curie temperature are substantially higher than room temperature (please refer to Figure 4 and later discussions). We assume it to be a sensible estimation to use the magnetic asymmetry of 10nm Fe on a vicinal substrate (about 16%) as a measure of M_0 for the 0.9nm Fe on the vicinal substrate. We also take into account the 8% difference between the films on vicinal and singular substrate for the normalisation of the singular results. The normalised data, as an approximate measure of the normalised spontaneous magnetisation, are then superimposed to the inset of Figure 3. It appears that our data are clustered close to the J=1/2 curve, just like that of bulk Fe and Ni, suggesting that the electron spin is likely to be the main contribution to the atomic moment. The larger deviation as the temperature approaches to the transition temperature is usually accounted for as the failure of the theory close to the critical point. We wish to emphasise that the above discussion depends sensitively on the estimation of M_0. To fit to the J=8 curve, a value of the magnetic asymmetry of about 25% would be required.

We shall now discuss the sensitivity of the Curie temperature with regard to additional Fe coverage. In Figure 4, the XMLD data taken close to room temperature are presented for the 0.9nm Fe on singular GaAs (Figure 3(a), 283K) and that for a further deposition of 0.35 nm Fe (Figure 3(b), 298K). Whilst the former showed an essentially zero magnetic asymmetry, the latter displayed a non-zero value. It is clear

from the results that the additional coverage of 0.35 nm of Fe has enhanced the magnetic interaction between the Fe atoms, resulting an increase of the Curie point to well above room temperature.

Our measurements suggest that in the initial stage of Fe coverage, the magnetic interaction has been weakened by inter-diffusion as well as islanding. Consequently the Curie temperature has been reduced to well below room temperature. There seems to be a critical Fe coverage, likely to correspond to a reasonable degree of coalescence of islands, above which the films become ferromagnetic at room temperature. However it does not seem to be clear that the non-ferromagnetic phase is of superparamagnetic in nature, especially considering the close proximity of the Fe islands and the relatively long range of the exchange interaction.

To summarise, we have carried out a temperature dependent study of the XMLD of ultrathin Fe films on singular and vicinal GaAs(001) substrates. Values of Curie temperature well below room temperature have been found, e.g., 240K for a 0.9 nm Fe on GaAs(001), but the values are highly sensitive to the stoichiometry and morphology of substrate surfaces as well as Fe overlayer coverage. We believe that this work demonstrates that as a surface-sensitive, element-specific probe of sample magnetisation, XMLD and its temperature dependent behaviours have tremendous potential in the study of magnetic characteristics of ultrathin films and related systems. Further work is planned to enable a more quantitative investigation.

Reference:

[1] I. Malajovich, J.J. Berry, N. Samarth, D.D. Awschalom: Nature **411**, 770 (2001)

[2] Y. Ohno, D.K. Young, B. Beschoten, F. Matsukura, H. Ohno, D.D. Awschalom: Nature **402**, 790 (1999)

[3] A.T. Filip. B.H. Hoving, F.J. Jedema, B.J. van Wees, B. Dutta, S. Borghs: Phys. Rev. B **62**, 9996 (2000)

[4] P.R. Hammar, B.R.Bennett, M.J. Yang, M. Johnson: Phys. Rev. Lett. **83**, 203 (1999)

[5] J.M. Kikkawa, D.D. Awschalom: Nature **397**, 139 (1999)

[6] J.M. Kikkawa, D.D. Awschalom: Phys. Rev. Lett. **80**, 4313 (1998)

[7] J.R. Waldrop and R.W. Grant, Appl. Phys. Lett. **34**, 630 (1979)

[8] G.A. Prinz, in *Ultrathin Magnetic Structures* edited by B. Heinrich and J.A.C. Bland (Spinger-Verlag, Berlin, 1994) Vol.2, pp 1-44

[9] A. Filipe, A. Schuhl, P. Galtier, Appl. Phys. Lett. **70**, 129 (1997); A. Filipe and A. Shuhl, J. Appl. Phys. **81**, 4359 (1997)

[10] Y.B. Xu, E.T. Kernohan, D.J. Freeland, A. Ercole, M. Tselepi and J.A.C. Bland, Phys. Rev. B **58**, 890 (1998)

[11] C.M. Gurtler, Y.B. Xu, J.A.C. Bland, J. M.M.M. **226**, 655 (2001)

[12] T.Zhang, M.Spangenberg, D.Greig, N.Takahashi, T-H,Shen, J.A.D.Matthew, S.Comelius, M.Rendall, E.A.Seddon, Appl. Phys. Lett. **78**, 961 (2001)

[13] N. Takahashi, T. Zhang, M. Spangenberg, D. Greig, T.H. Shen, S. Cornelius, E.A. Seddon and J.A.D. Matthew, Surf. Rev. and Lett. **9**, 693 (2002)

[14] F.O. Schmann, R.F. Willis, K.G. Goodman and J.G. Tobin, Phys. Rev. Lett. **79**, 5166 (1997)

[15] F.O. Schmann, R.F. Willis, and J.G. Tobin, J.V.S.T. **A18,** 1259 (2000)

[16] J.G. Tobin and F.O. Schumann, Surf. Sci. **478**, 211(2001)

[17] F.J. Himpsel, Phys. Rev. Lett. **67**, 2363 (1991)

[18] M. Hochstrasser, J.G. Tobin, E. Rotenberg and S.D. Kevan, Phys. Rev. Lett. **89**, 216802 (2002)

[19] S.A. Morton, G.D. Waddill, S. Kim, I.K. Schuller, S.A. Chambers, and J.G. Tobin, Surf. Sc. Lett. **513**, L451 (2002)

[20] D.A. Shirley, Phys. Rev. B, **5**, 4709 (1972)

[21] H.B. Rose, T. Kinoshita, C. Roth and F.U. Hillebrect, in Spin-orbit-Influenced Spectroscopies of Magnetic Solids, edited by G. Schutz and H. Ebert (Springer, Berlin, 1996) p105

[22] G.D. Waddill, J.G. Tobin and D.P. Pappas, Phys. Rev. B, Rapid Comm. **46**, 552 (1992)

[23] F. Tyler, Phil. Mag., **11**, 596 (1931)

Caption:

Figure 1. Dichroism in Fe 2p core level photoemission of 2.7nm thick Fe film on singular GaAs(001) substrate, as excited by 810 eV p-linearly polarised photons. 'Mag up' and 'Mag down' indicate the two remnant states parallel and antiparallel to [110] direction.

Figure 2. Dichroism in Fe 3p core level photoemission of 2.7nm thick Fe film on singular GaAs(001) substrate, with 160 eV p-linearly polarised photons. The meaning of the 'Mag up' and 'Mag down' notations is the same as in Figure 1. A_{TOT} denotes the peak-to-peak asymmetry.

Figure 3. A temperature dependence of the peak-to-peak magnetic asymmetry ('Total Asymmetry') of Fe 3p photoelectrons for 0.9nm Fe films on GaAs (001) substrates. The inset shows a likely fit to the temperature dependent spontaneous magnetisation as predicted by the Weiss-type theory [23].

Figure 4. Dichroism measurements of Fe 3p close to room temperature on (a) 0.9nm Fe on singular GaAs (001) and (b) followed by an additional 0.35nm Fe deposition. The meaning of the 'Mag up' and 'Mag down' notations is the same as in Figure 1.
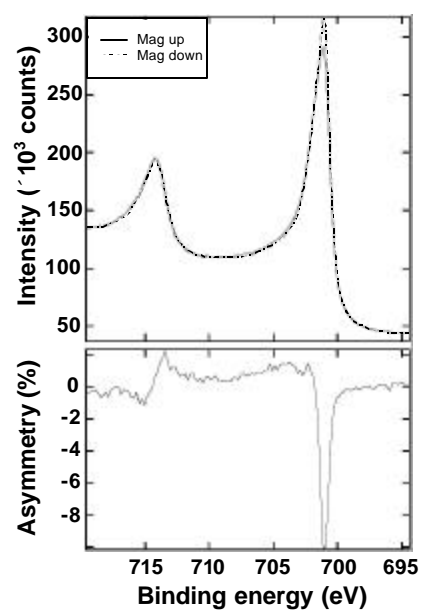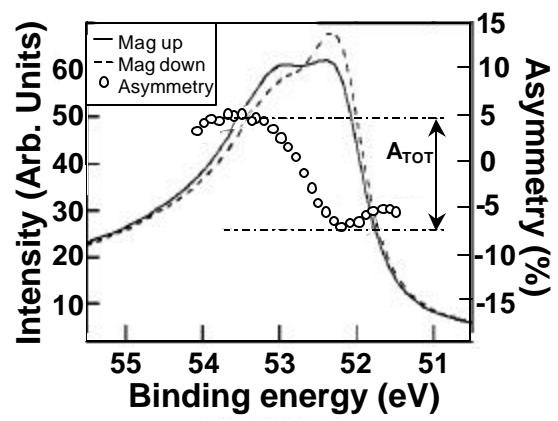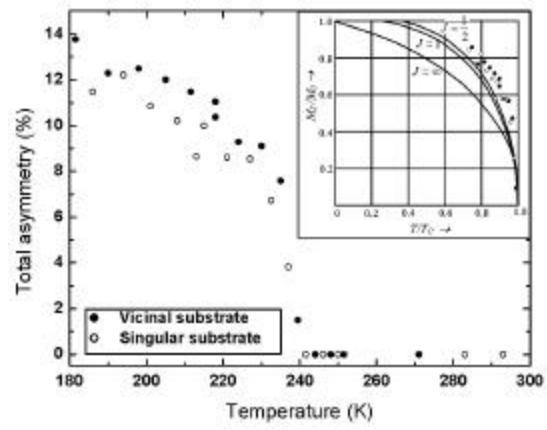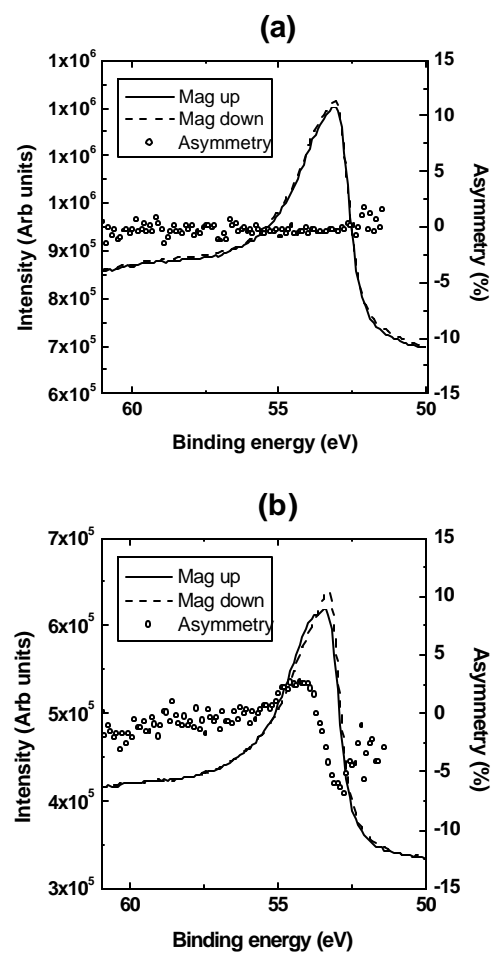
Figure 1

Figure 2

Figure 3

(a)

(b)

Figure 4